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(54) Monomeric direactive mesogenic compounds, intermediates therefor and polymers therefrom

(57) The invention relates to direactive mesogenic compounds or mixtures thereof comprising mesogen-containing molecule, said mesogens having two side chains attached thereto which contain a terminal polymerizable functional group, said mesogens and said functional groups being separated by at least two to twenty spacer atoms, wherein both spacer groups have different chain length.

The invention is directed to monomeric direactive mesogenic compounds as defined in claims 1, 2 and 7; polymers prepared by polymerising such monomers (claim 9); and intermediated (as defined in claim 10) for such monomers.

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Direactive mesogenic compound

The invention relates to

5 direactive mesogenic compounds or mixtures thereof obtainable by

a) treating a mesogenic diol of formula I,



10

in which

MG is a mesogenic group.

15 with a mixture of the halides of formula II and III.



20

in which

X¹ and X² are each independently Cl, Br or I

25 m and n are different integers between 1 and 20

R^a and R^b are each independently groups selected from



30

wherein

W is H, CH₃ or Cl.

35 in the presence of a base, and

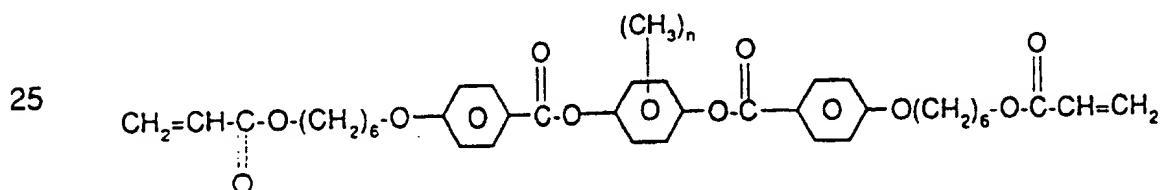
b) treating the resulting intermediate

- in the case of R^a and R^b being -CH₂OH, with a vinyl derivative of formula CH₂=CW-(CO)_a-O- or a reactive derivative thereof, in which a is 0 or 1
 - in the case of R^a and R^b being -CH=CWH with a perbenzoic acid.

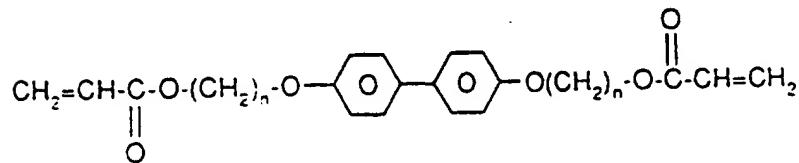
10 The invention furthermore relates to the preparation of such compounds
and to their use in electrooptical scattering systems and for the
preparation of oriented liquid crystal polymers.

15 Reactive liquid crystal compounds can be polymerized in situ, whilst in
their liquid crystal phase, to give highly crosslinked anisotropic films which
can be used, for example, as polarizing beam splitters (see, for example,
EP 0,428,213). Reactive liquid crystal compounds have furthermore been
proposed for electrooptical scattering systems (see, for example,
EP 0,451,905), cholesteric polarizers (e.g. EP 0,606,940) and
compensation films for STN displays (e.g. EP 0,423,881).

Reactive liquid crystal diesters of formula



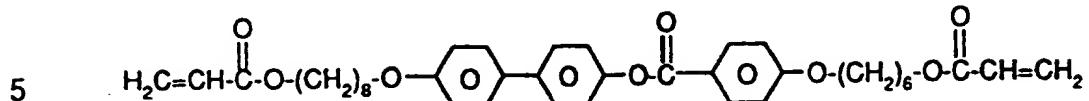
are mentioned in EP 0,261,712 (n=0), EP 0,331,233 (n=1). Reactive liquid crystal biphenyls of formula



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are disclosed by EP 0,405,713.

The International Patent application WO 93/22397 discloses a compound of formula



These reactive liquid crystalline compounds often exhibit, however, rather high melting points disadvantageous values of the birefringence and comparable narrow mesophase ranges.

10 In view of the broad range of applications of reactive liquid crystal compounds it was desirable to have available further compounds of this type which fulfill the various requirements such as a reasonably low melting point, a high birefringence, a broad mesogenic range and
15 preferably an enantiotropic nematic range to a high degree.

20 It was an object of the present invention to provide new reactive liquid crystalline compounds with advantageous properties thus extending the pool of reactive liquid crystal compounds available to the expert. Other objects of the present invention can be taken from the following detailed specification.

25 The present invention thus relates to reactive mesogenic compounds or mixtures thereof obtainable by treating mesogenic diols of formula I, in particular those having a symmetric structure unit with a mixture of halides of formula II and III and to their use in electrooptical systems of scattering type and for the preparation of oriented liquid crystal polymers. The invention furthermore relates to the preparation of compounds according to formula I.
30

Preferred embodiments of the present invention are:

- 35 a) Composition of dieractive compounds comprising at least one compound of each formula IV, V and VI,



V



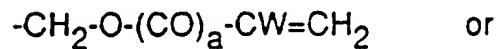
VI

5

in which MG, m and n have the meaning given, and

R¹ and R² are each independently

10



where a and W have the meaning given.

b) Diereactive compound or mixture thereof

15

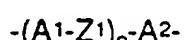
in which

m - n ≥ 1, in particular 2, 3 or 4.

c) Diereactive compound or mixture thereof in which

20

MG is a mesogenic group of formula VII,



VII

25

in which

A¹ and A² are each independently

30

(a) 1,4-phenylene in which one or two CH groups may be replaced by N;

(b) 1,4-cyclohexylene in which one or two non-adjacent CH₂ groups may be replaced by -O- or one -CH- group may be replaced by -C(CN)-;

35

(c) naphthalene-2,6-diyl;

it being possible that group (a) is substituted by halogen
cyano or alkyl, alkoxy or alkanoyl with 1 to 6 C atoms,

5

Z¹ is each independently -COO-, -O-CO-, -CH₂-CH₂-,
-C≡C-, -CH₂O-, -OCH₂- or a single bond, and

o is 1, 2 or 3.

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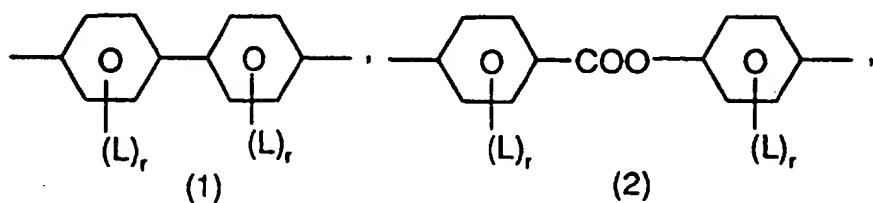
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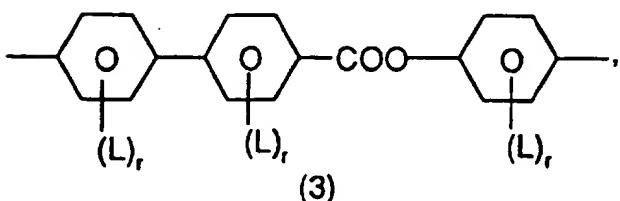
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d) Direactive compound or mixture thereof in which MG is selected from the structure elements (1) to (6).

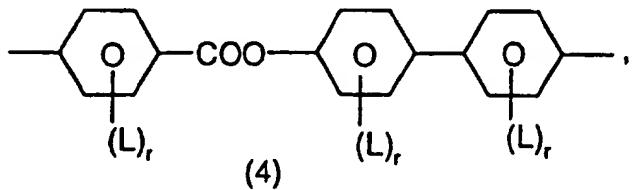
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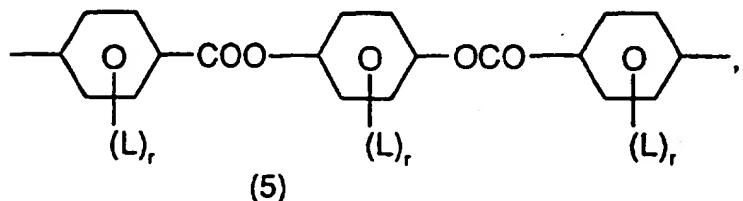
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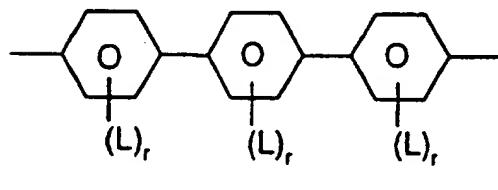
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20



25



(6)

in which

30

L is CH₃, Cl, F, OCH₃ or -CO-CH₃, and

r is 0, 1, 2 or 4.

35

e) Direactive compound or mixture thereof in which

n and m are given by the following table:

5	m	5	5	5	4	4
	n	2	3	4	2	3

A further aspect of the present invention are direactive compounds of
formula IV A

10



in which

15

R¹ and R² have the meaning given,

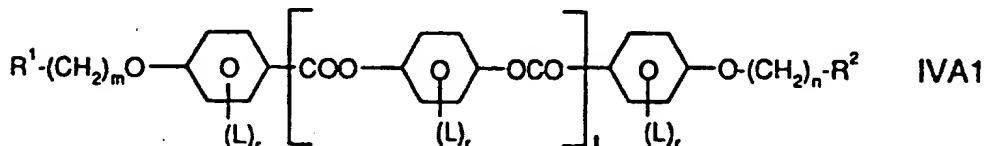
m and n are different integers between 2 and 10, and

20

MG is a mesogenic group, the core of which being symmetrical,
preferably a structure element of formula (1), (5) or (6), in
particular

direactive compounds of the formula IVA1

25



30

in which

R¹, R²,
L and r have the meaning given,

35

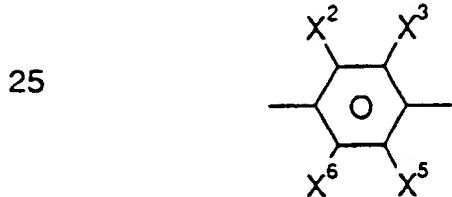
m and n are different integers between 2 and 10, and

t is 0 or 1.

5 Another aspect of the invention are the polymers prepared by polymerizing a monomer according to any of the preceding claims and
10 chemical intermediate compounds or mixtures thereof useful in preparing direactive compounds or mixtures thereof according to any of the Claim 1 to 8, comprising mesogen-containing molecules, said mesogens having two side chains attached thereto which contain hydroxyl or vinyl group at the end thereof, said mesogens and said functional groups being separated by at least two to twenty spacer atoms, wherein both spacer
15 groups have different chain length.

Above and below, the term reactive mesogenic compounds refers to reactive rod-like molecules which may be enantiotropic, monotropic or isotropic, preferably, however, enantiotropic or monotropic.

20 In the inventive compounds in which MG is a mesogenic group of formula VII, A¹ and A² can be independently from each other an unsubstituted or a substituted 1,4-phenylene group of formula

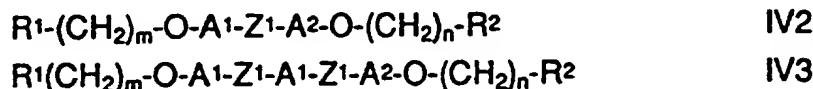


30 X², X³, X⁵ and X⁶ can be independently from each other H, F, Cl, methyl or CN.

In the following, for the sake of simplicity, the following notation will be used:

Phe. 2 X² 3 X³ 5 X⁵ 6 X⁶ is a 1,4-phenylene group carrying in 2-position the group X², in 3-position the group X³ etc.; in case X², X³, X⁵ and/or X⁶, denote H, this will not be specified in above notation, i.e. only true substitutions will be listed. Thus Phe, for example, is an unsubstituted 1,4-phenylene group while Phe.2F 5 Cl is a 2-fluoro-5-chloro-1,4-phenylene group. Furthermore, Pyr is pyrimidine-2,5-diyl, Pyd is pyridine -2,5-diyl and Nap is a naphthalene-2,6-diyl group. The notation Pyr and Pyd in each case include the 2 possible positional isomers.

- 10 The compounds according to formula IV comprise 2- and 3-ring compounds (n=1 or 2) of formula IV2 and IV3:



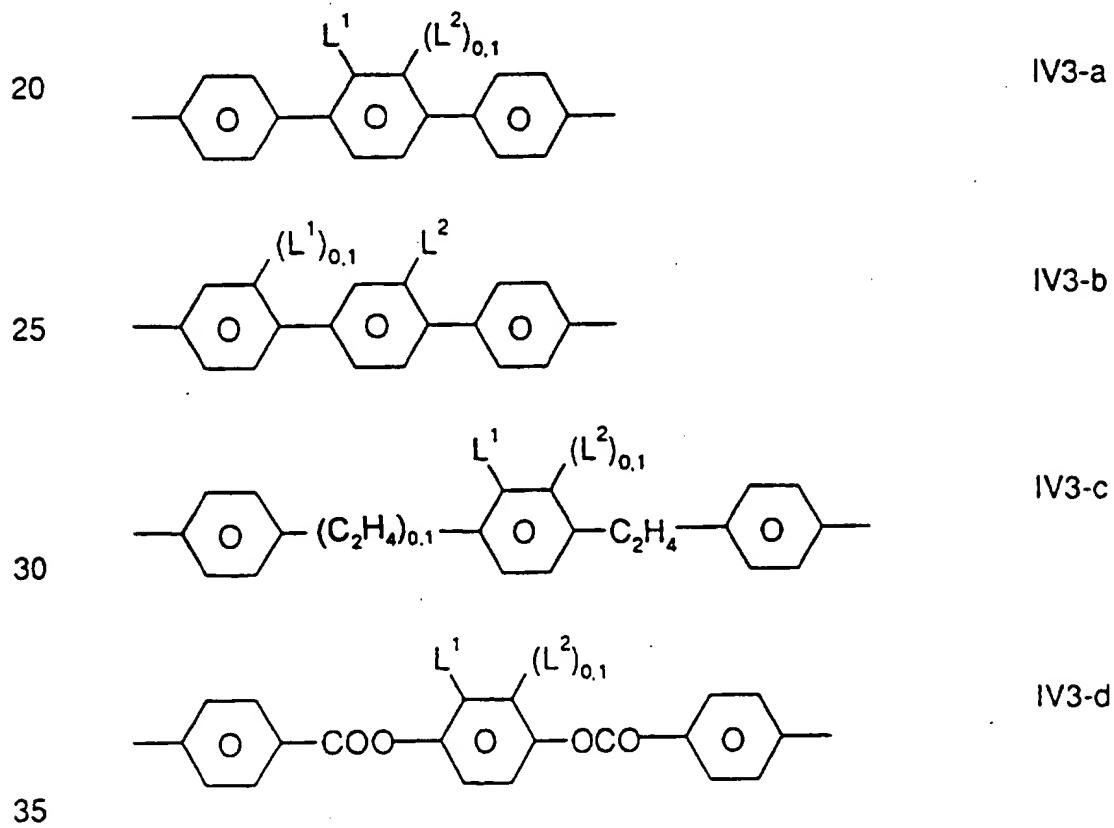
- 15 In the 3-ring compounds of formula IV3, the ring groups A¹ can be chosen independently from each other.

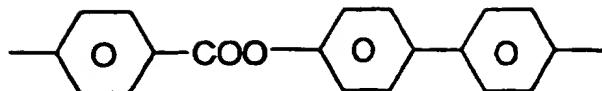
- 20 Especially preferred is a smaller group of 2-ring compounds exhibiting the following structures for -A¹-Z¹-A²-:

	-Phe.2CH ₃ -Phe-	IV2-1
	-Phe.3CH ₃ -Phe-	IV2-2
	-Phe.2Cl-Phe-	IV2-3
25	-Phe.3Cl-Phe-	IV2-4
	-Phe.2CN-Phe-	IV2-5
	-Phe.3CN-Phe-	IV2-6
	-Phe.2Cl3Cl-Phe-	IV2-7
	-Phe.2Cl3F-Phe-	IV2-8
30	-Phe.2F-Phe-	IV2-9
	-Phe.3F-Phe-	IV2-10
	-Phe.-Phe-	IV2-11
	-Phe.F-Nap-	IV2-12
	-Phe.2Cl-Nap-	IV2-13

	-Phe.F-Nap-	IV2-14
	-Phe.3Cl-Nap-	IV2-15
	-Phe.2F-Pyr-	IV2-16
	-Phe.2F-Pyr-	IV2-17
5	-Phe.2CH ₃ -Pyd-	IV2-18
	-Phe.2Cl-Pyd-	IV2-19
	-Phe.F-CH ₂ CH ₂ -Phe-	IV2-20
	-Phe.3F-CH ₂ CH ₂ -Phe-	IV2-21
	-Phe.2Cl-CH ₂ CH ₂ -Phe-	IV2-22
10	-Phe.3Cl-CH ₂ CH ₂ -Phe-	IV2-23
	-Phe.2CN-CH ₂ CH ₂ -Phe-	IV2-24
	-Phe.3CN-CH ₂ CH ₂ -Phe-	IV2-25
	-Phe.2Cl3Cl-CH ₂ CH ₂ -Phe-	IV2-26
	-Phe.2Cl3F-CH ₂ CH ₂ -Phe-	IV2-27
15		

The 3-ring compounds according to formula IV3 preferably exhibit the following structures for -A₁-Z₁-A₁-Z₁-A₂:





IV3-e

- 5 In these structures, IV3-a to IV3-d, L¹ and L² denote independently from each other H, -Cl, -F, -CN and C_rH_{2r+1-s} and, in particular, -Cl, -F, -CN, -CH₃, and/or -C₂H₅.

10 Especially preferred are the following patterns:

	-Phe-Phe.2CH ₃ -Phe-	IV3-a-1
	-Phe-Phe.2Cl-Phe-	IV3-a-2
	-Phe-Phe.2CN-Phe-	IV3-a-3
15	-Phe-Phe.2F-Phe-	IV3-a-4
	-Phe-Phe-Phe-	IV3-a-5
	-Phe-Phe.2C ₂ H ₅ -Phe-	IV3-a-6
	-Phe-Phe.3CH ₃ -Phe-	IV3-b-1
20	-Phe-Phe.3Cl-Phe-	IV3-b-2
	-Phe-Phe.3CN-Phe-	IV3-b-3
	-Phe-Phe.3F-Phe-	IV3-b-4
	-Phe-Phe.3C ₂ H ₅ -Phe-	IV3-b-5
25	-Phe.3F-Phe.3Cl-Phe-	IV3-b-6
	-Phe.3F-Phe.3CH ₃ -Phe-	IV3-b-7
	-Phe.3Cl-Phe.3Cl-Phe-	IV3-b-8
	-Phe.3Cl-Phe.3CH ₃ -Phe-	IV3-b-9
	-Phe-Phe.2Cl-Phe.3Cl-	IV3-b-10
30	-Phe-Phe.3Cl-Phe.3Cl-	IV3-b-11
	-Phe-Phe.2Cl-Phe.2Cl-	IV3-b-12
	-Phe-Phe.3Cl-Phe.2Cl-	IV3-b-13
	-Phe-Phe.2CH ₃ -Phe.3Cl-	IV3-b-14
	-Phe-Phe.3CH ₃ -Phe.3Cl-	IV3-b-15
35	-Phe-Phe.2CH ₃ -Phe.2Cl-	IV3-b-16

	-Phe-Phe.3CH ₃ -Phe.2Cl-	IV3-b-17
	-Phe-Phe.2F-Phe.3Cl-	IV3-b-18
	-Phe-Phe.3F-Phe.3Cl-	IV3-b-19
	-Phe-Phe.2F-Phe.2Cl-	IV3-b-20
5	-Phe-Phe.3F-Phe.2Cl-	IV3-b-21
	-Phe-Phe.2Cl-Phe.3CN-	IV3-b-22
	-Phe-Phe.3Cl-Phe.3CN-	IV3-b-23
	-Phe-Phe.2Cl-Phe.2CN-	IV3-b-24
	-Phe-Phe.3Cl-Phe.2CN-	IV3-b-25
10	-Phe-Phe.2CH ₃ -Phe.3CN-	IV3-b-26
	-Phe-Phe.3CH ₃ -Phe.3CN-	IV3-b-27
	-Phe-Phe.2CH ₃ -Phe.2CN-	IV3-b-28
	-Phe-Phe.3CH ₃ -Phe.2CN-	IV3-b-29
	-Phe-Phe.3F-Phe.3CN-	IV3-b-30
15	-Phe-Phe.2F-Phe.3CN-	IV3-b-31
	-Phe-Phe.3F-Phe.2CN-	IV3-b-32
	-Phe-Phe.2F-Phe.2CN-	IV3-b-33
	-Phe-Phe.2F-Phe.2F-	IV3-b-34
	-Phe-Phe.3F-Phe.3F-	IV3-b-35
20	-Phe-Phe.2CH ₃ -C ₂ H ₄ -Phe-	IV3-c-1
	-Phe-Phe.2Cl-C ₂ H ₄ -Phe-	IV3-c-2
	-Phe-Phe.2CN-C ₂ H ₄ -Phe-	IV3-c-3
	-Phe-Phe.2F-C ₂ H ₄ -Phe-	IV3-c-4
25	-Phe-Phe.2C ₂ H ₅ -C ₂ H ₄ -Phe-	IV3-c-5
	-Phe-Phe.2Cl3F-C ₂ H ₄ -Phe-	IV3-c-6
	-Phe-Phe.2Cl3Cl-C ₂ H ₄ -Phe-	IV3-c-7
	-Phe-C ₂ H ₄ -Phe.2CH ₃ -C ₂ H ₄ -Phe-	IV3-c-8
	-Phe-C ₂ H ₄ -Phe.2Cl-C ₂ H ₄ -Phe-	IV3-c-9
30	-Phe-C ₂ H ₄ -Phe.2CN-C ₂ H ₄ -Phe-	IV3-c-10
	-Phe-C ₂ H ₄ -Phe.F-C ₂ H ₄ -Phe-	IV3-c-11
	-Phe-C ₂ H ₄ -Phe.2OCF ₃ -C ₂ H ₄ -Phe-	IV3-c-12
	-Phe-COO-Phe.OCO-Phe-	IV3-d-1
35	-Phe-COO-Phe.2CH ₃ -OCO-Phe-	IV3-d-2

	-Phe-COO-Phe.3CH ₃ -OCO-Phe-	IV3-d-3
	-Phe-COO-Phe.2CH ₃ 3CH ₃ -OCO-Phe-	IV3-d-4
	-Phe-COO-Phe.2OCH ₃ -OCO-Phe-	IV3-d-5
	-Phe-COO-Phe.2Cl-OCO-Phe-	IV3-d-6
5	-Phe-COO-Phe.2F-OCO-Phe-	IV3-d-7
	-Phe-COO-Phe.2F3F-OCO-Phe-	IV3-d-8
	-Phe-COO-PhePhe-	IV3-e-1
	-Phe-COO-Phe.2FPhe-	IV3-e-2
10	-Phe-COO-Phe.3FPhe-	IV3-e-3
	-Phe-COO-PhePhe.2F-	IV3-e-4
	-Phe-COO-PhePhe.3F-	IV3-e-5
	-Phe.2F-COO-PhePhe-	IV3-e-6
	-Phe.3F-COO-PhePhe-	IV3-e-7

15

It was observed that the stability of 3-ring compounds wherein one of the 2 groups Z¹ is -COO- or -OCO- while the other denotes a single bond, can be increased if the compound is laterally di- or higher substituted, particularly di-substituted by -Cl, -F, -CN and/or -CH₃. Compounds of this type are preferred.

20

Especially preferred are further 3-ring compounds where both groups Z¹ are either -COO-, or -OCO- and at least one of the rings A¹, A^{1'} and A² are at least mono substituted.

25

In the compounds of formula IV R¹ is CH₂ = CW-COO-CH₂-, CH₂ =



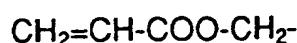
CH-O-CH₂-, HW-C-C-, with W being H, Cl or alkyl with 1-5 C atoms and

30

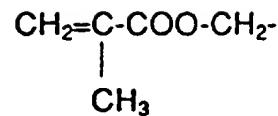
m being 1-7.

Preferably, R¹ and R² is a vinyl group, an acrylate group, an epoxy group and especially preferred are the following means of R¹ and R²:

35

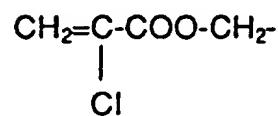


R¹-1



R1-2

5

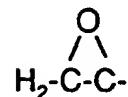


R1-3



R1-4

10



R1-5

with alkyl denoting C₁-C₃-alkyl and m being 1-5.

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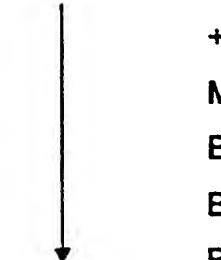
The reaction methods mentioned are briefly summarized in the following synthetic tree:

Scheme 1

5

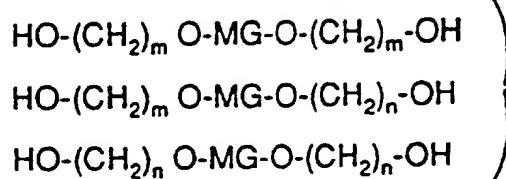


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Mixture of
 $\text{Br-(CH}_2\text{)}_m\text{-OH}$
 $\text{Br-(CH}_2\text{)}_n\text{-OH}$
Butanone, K_2CO_3 , heat

15



} Mixture

20

Butylvinyl ether
DCM
1,10-Phenanthroline
Pd (II)-Acetate



25

Mixture of
"Divinylethers"

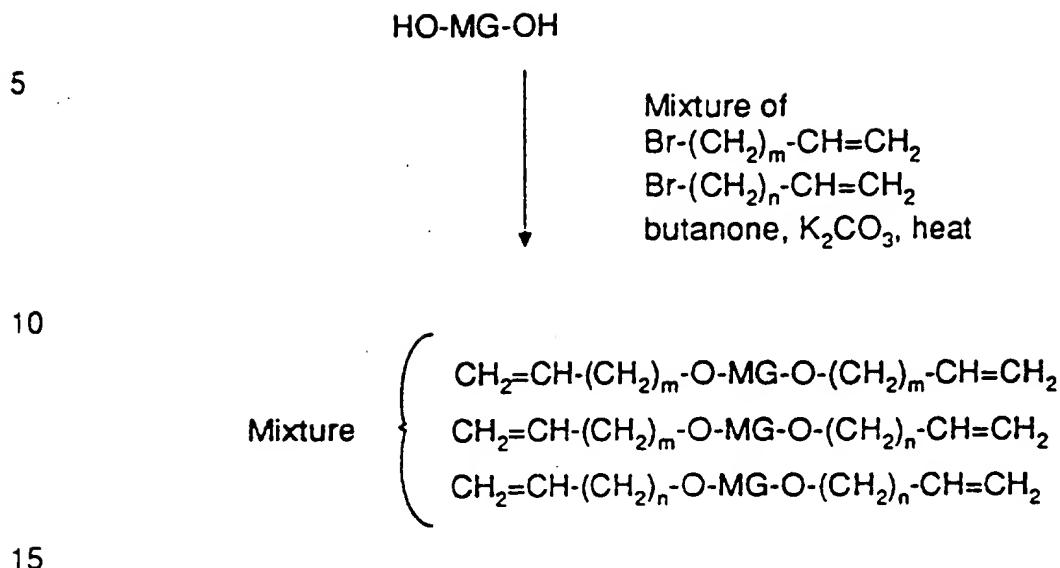
Mixture of
"diacrylates"

DCM = dichloromethane

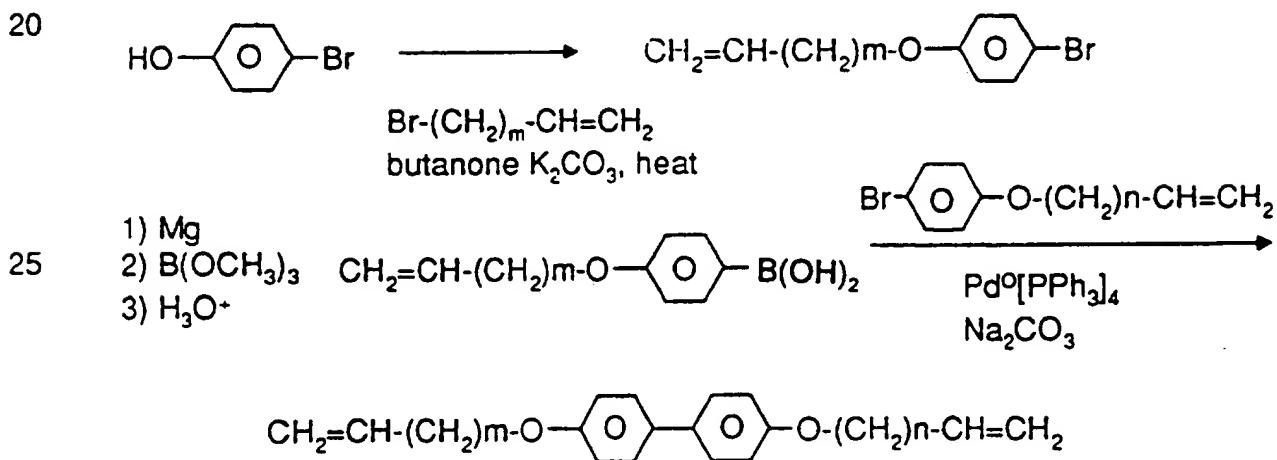
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Scheme II



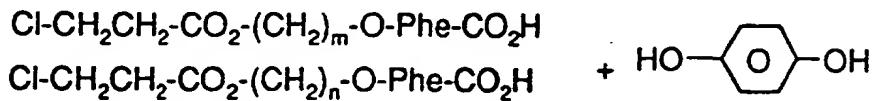
Scheme III



Certain compounds or mixtures according to the invention are obtainable in a "one-pot-synthesis" as outlined in scheme IV:

5 Scheme IV

10



1) TFAA/DCM
2) TEA/DCM

15



20



TFAA = trifluoroacetic acid

TEA = triethylamine

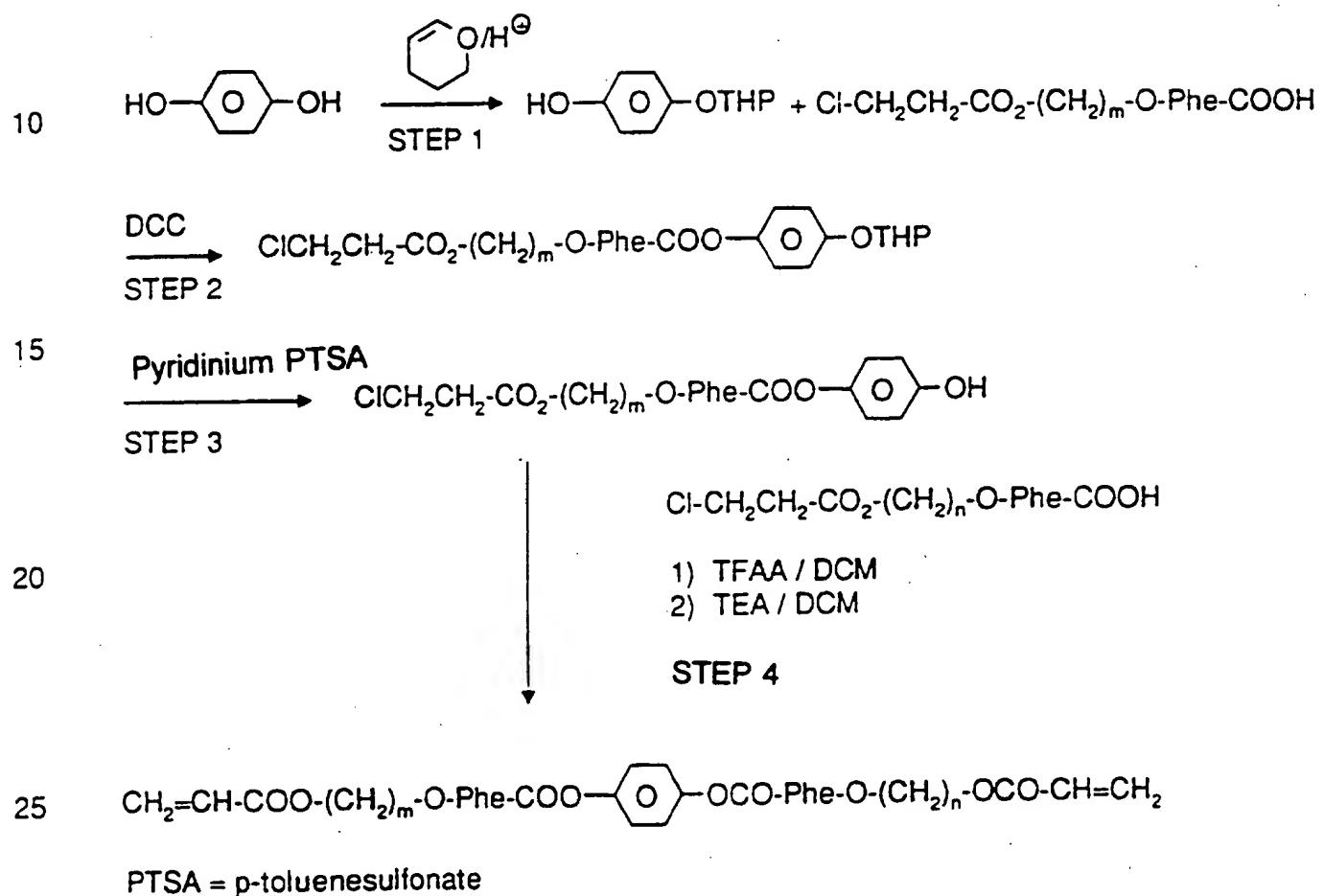
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Individual compounds of this type can be obtained according to schemes V to VII:

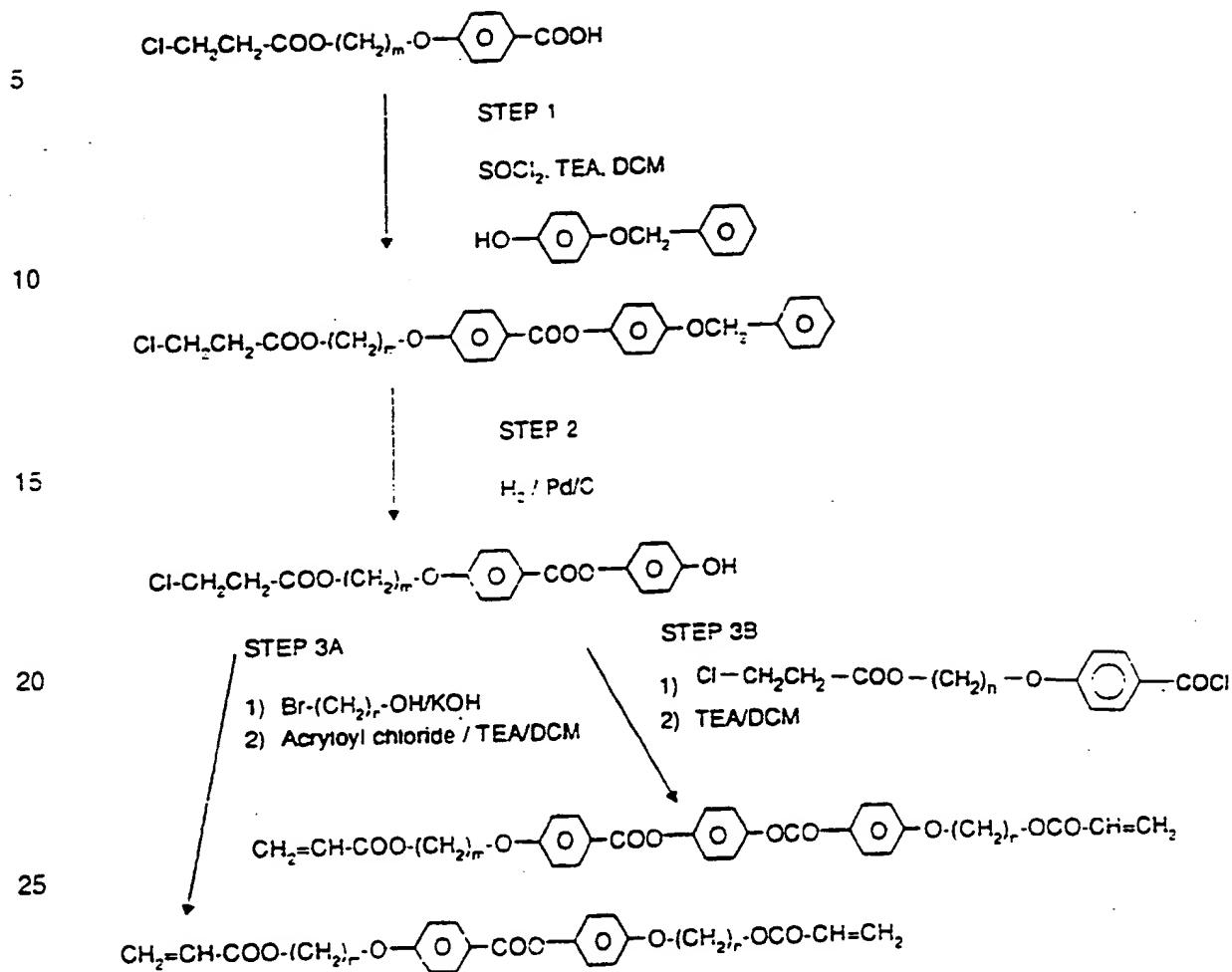
5 Scheme V



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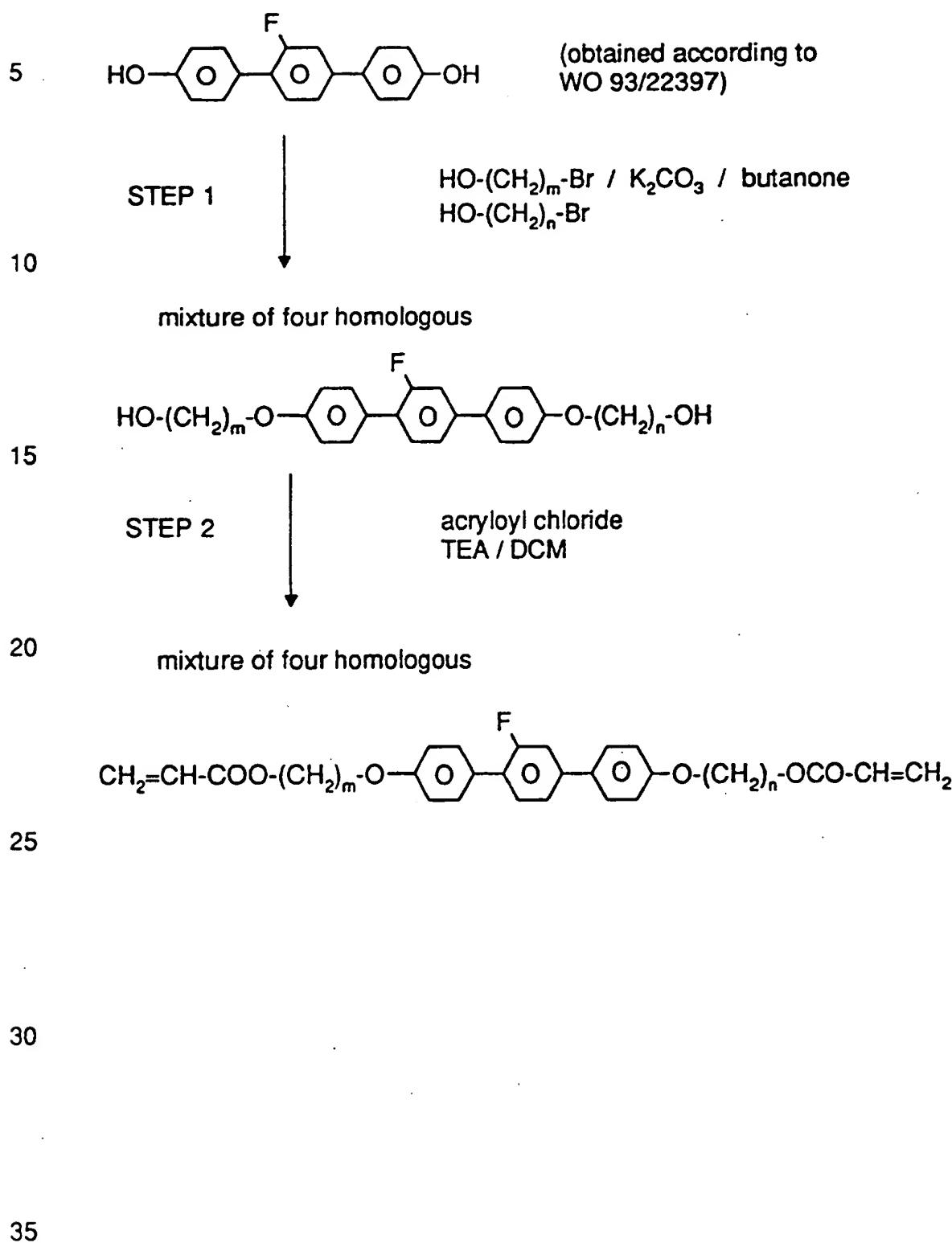
Scheme VI



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Scheme VII



The reaction schemes mentioned above are to illustrate the invention without restricting it. The expert can choose other reaction methods without any inventive efforts.

- 5 In the following and in the preceding, all percentages given are percentages by weight. Temperatures are given in degrees Celsius.

The following examples are intended to illustrate the invention without restricting it.

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Example 1

The reactive liquid crystalline compound (1)

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is prepared via the sequence of reaction steps shown in scheme V. In step 4 of scheme V 2.2 moles TFAA are added dropwise to a solution 2 moles of the phenol obtained in step 3 of scheme V and 2 moles of the benzoic acid in 2 l of DCM.

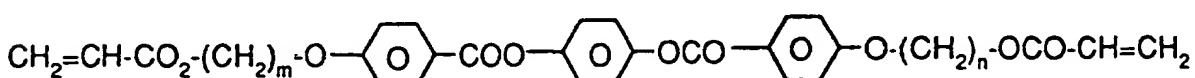
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The reaction mixture is stirred at room temperature for 16 hours. Then 5 moles of TEA in 1 l of DCM are added. The mixture is stirred for 16 hours.

Aqueous work-up and column chromatography give (1) which shows K 111 S.

The following compounds are obtained analogously:

30



Compound No.	m	n	phase transition temperatures (°C)
5	(2)	3	K 76 N 160 I
	(3)	4	K 67 N 165 I
	(4)	4	K 78 S 82 N 159 I
	(5)	5	K 78 S 80 N 162 I
10	Comp 1	4	K 105 N 164 I
	Comp 2	5	K 91 N 167 I
	Comp 3	6	K 105 (S 95) 152 I

Example 2

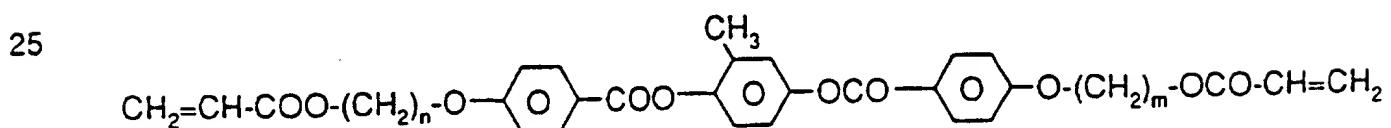
A mixture of 1 mol $\text{Cl}-\text{CH}_2\text{CH}_2\text{-COO-(CH}_2)_4\text{-O}-\text{C}_6\text{H}_4\text{-COOH}$,

15 1 mol $\text{Cl}-\text{CH}_2\text{CH}_2\text{-COO-(CH}_2)_6\text{-O}-\text{C}_6\text{H}_4\text{-COOH}$, and 1 mol

p-hydroquinone is treated with 2.2 moles of TFAA followed by 2.5 moles of TEA.

20 A mixture consisting of 1 part comp 1 and 1 part comp 3 and 2 parts of Compound No. (4) is obtained which shows a melting point of 56 °C and a clearing point of 163 °C.

Analogously a mixture of the following compounds is obtained:



	n	m	parts
30	3	3	1
	6	6	1
	3	6	1
	6	3	1

35 This composition shows a melting point below 30 °C and a clearing point of 119 °C.

Example 3

A mixture of 0.5 moles of biphenol, 1.3 moles of potassium hydroxide, 0.6 moles of 3-bromopropanol and 0.6 moles of 3 bromohexanol is heated

5 in 2 litres of butanone for 16 hrs. After aqueous work-up a mixture of

1 part of $\text{HO}-(\text{CH}_2)_3-\text{O}-\text{C}_6\text{H}_4-\text{C}_6\text{H}_4-\text{O}-(\text{CH}_2)_3-\text{OH}$

1 part of $\text{HO}-(\text{CH}_2)_6-\text{O}-\text{C}_6\text{H}_4-\text{C}_6\text{H}_4-\text{O}-(\text{CH}_2)_6-\text{OH}$, and

10 2 parts of $\text{HO}-(\text{CH}_2)_6-\text{O}-\text{C}_6\text{H}_4-\text{C}_6\text{H}_4-\text{O}-(\text{CH}_2)_3-\text{OH}$

is obtained.

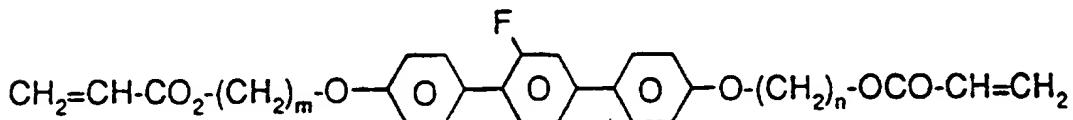
This mixture is treated with 1.2 moles of acryloyl chloride and 1.2 moles of TEA in 1.5 litres of dichloromethane DCM and refluxed for 3 hours.

15 After aqueous work-up the resulting reaction mixture is purified by column-chromatography to yield a mixture of diacrylates (6), (7) and (8).



Compound No.	m	n	parts
(6)	3	3	1
(7)	6	6	1
25 (8)	3	6	2

Analogously a mixture of compounds of formulae (9), (10), (11) and (12) is obtained



	<u>Compound No.</u>	<u>m</u>	<u>n</u>	<u>parts</u>
5	(9)	3	3	1
	(10)	6	6	1
	(11)	3	6	1
	(12)	6	3	1

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Claims

1. Direactive mesogenic compounds or mixtures thereof obtainable
by

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a) treating a mesogenic diol of formula I.



(I)

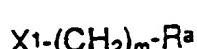
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in which

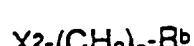
MG is a mesogenic group,

15

with a mixture of the halides of formula II and III,



(II)



(III)

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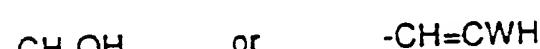
in which

X¹ and X² are each independently Cl, Br or I

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m and n are different integers between 1 and 20

R^a and R^b are each independently groups selected from



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wherein

W is H, CH₃ or Cl.

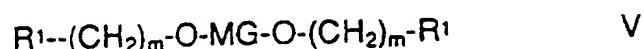
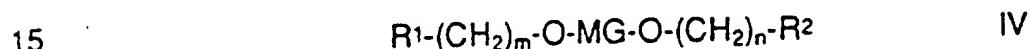
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in the presence of a base, and

b) treating the resulting intermediate

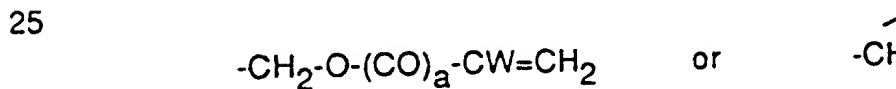
- 5 - in the case of R^a and R^b being -CH₂OH, with a vinyl derivative of formula CH₂=CW-(CO)_a-O- or a reactive derivative thereof, in which a is 0 or 1
- 10 - in the case of R^a and R^b being -CH=CWH with a perbenzoic acid.

2. Composition of direactive compounds comprising at least one compound of each formula IV, V and VI,



20 in which MG, m and n have the meaning given, and

R¹ and R² are each independently



wherein a and W have the meaning given.

3. Direactive compound or mixture thereof according to claim 1 or 2

30 in which

m - n ≥ 1.

4. Directive compound or mixture thereof according to any of the preceding claim in which

MG is a mesogenic group of formula VII,

$$5 \quad -(A^1 - Z^1)_0 - A^2 - \quad VII$$

in which

10 A₁ and A₂ are each independently

- (a) 1,4-phenylene in which one or two CH groups may be replaced by N;
 - (b) 1,4-cyclohexylene in which one or two non-adjacent CH₂ groups may be replaced by -O- or one -CH- group may be replaced by -C(CN)-;
 - (c) naphthalene-2,6-diyl;

20 it being possible that group (a) is substituted by halogen cyano or alkyl with 1 to 6 C atoms,

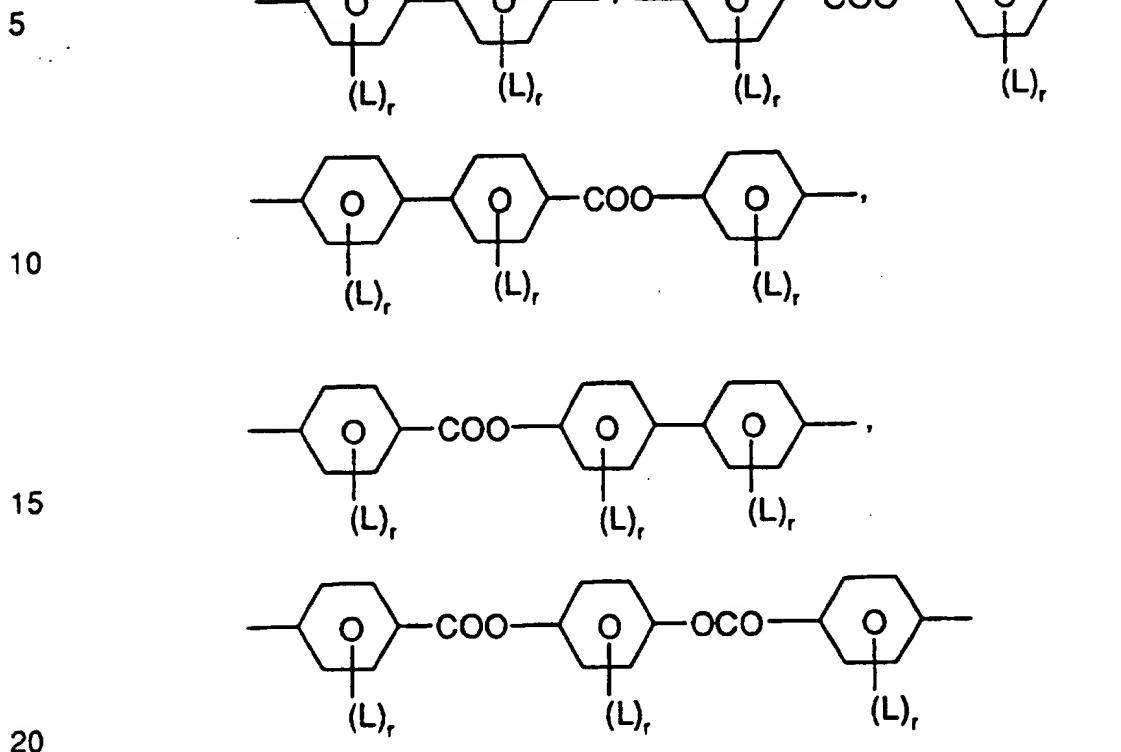
Z^1 is each independently -COO-, -O-CO-, -CH₂-CH₂-, -C≡C-, -CH₂O-, -OCH₂- or a single bond, and

is 1, 2 or 3.

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5. Directive compound or mixture thereof according to any of the preceding claims in which MG is selected from



in which

L is CH_3 , Cl, F, OCH_3 or $-\text{CO-CH}_3$, and

6. Directive compound or mixture thereof according to any of the preceding claims in which

30 n and m are given by the following table:

m	5	5	5	4	4
n	2	3	4	2	3

7. Direactive compound of formula IV A



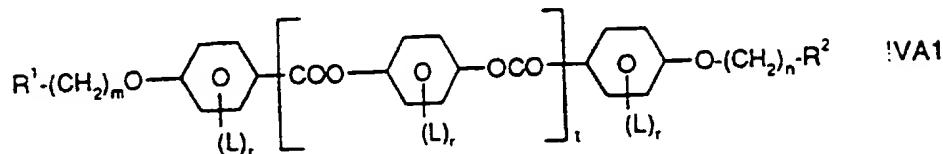
5 in which

R¹ and R² have the meaning given,

m and n are different integers between 2 and 10, and

10 MG is a mesogenic group, the core of which being
symmetrical.

8. Direactive compound according to claim 7 of the formula IVA1



15 in which

20 R¹, R²,
L and r have the meaning given,

m and n are different integers between 2 and 10, and

25 t is 0 or 1.

9. Polymers prepared by polymerizing a monomer according to any
of the preceding claims.

30 10. Chemical intermediate compounds or mixtures thereof useful in
preparing direactive compounds or mixtures thereof according to
any of Claims 1 to 8, comprising mesogen-containing molecules,
35 said mesogens having two side chains attached thereto

which contain hydroxyl or vinyl group at the end thereof, said mesogens and said functional groups being separated by at least two to twenty spacer atoms, wherein both spacer groups have different chain length.

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31

Application No: GB 9502294.3
Claims searched: 1-10

Examiner: Stephen Quick
Date of search: 26 April 1996

Patents Act 1977
Search Report under Section 17

Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.O): (not searched)

Int Cl (Ed.6): (not searched)

Other: Online: CAS ONLINE

Documents considered to be relevant:

Category	Identity of document and relevant passage	Relevant to claims
X	WO93/22397 A1 (MERCK PATENT), see compounds 4.1 (page 68; re claim 1) and 3.10 (page 67; re claim 10); acknowledged in this application	1 & 10 at least
X	Chem. Abs. 123:143045	1 & 10 at least
X	Chem. Abs. 122:33370	1 & 10 at least
X	Chem. Abs. 121:302740	1 & 10 at least
X	Chem. Abs. 121:256718	1 & 10 at least
X	Chem. Abs. 120:324302	1 & 10 at least
X	Chem. Abs. 120:272840	1 & 10 at least
X	Chem. Abs. 120:272839	1 & 10 at least
X	Chem. Abs. 120:165617	1 & 10 at least

X Document indicating lack of novelty or inventive step
Y Document indicating lack of inventive step if combined with one or more other documents of same category.

A Document indicating technological background and/or state of the art.
P Document published on or after the declared priority date but before the filing date of this invention.
E Patent document published on or after, but with priority date earlier



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32

Application No: GB 9502294.3
Claims searched: 1-10

Examiner: Stephen Quick
Date of search: 26 April 1996

Category	Identity of document and relevant passage	Relevant to claims
X	Chem. Abs. 120:19350	1 & 10 at least
X	Chem. Abs. 119:227845	1 & 10 at least
X	Chem. Abs. 119:97676	1 & 10 at least
X	Chem. Abs. 118:30690	1 & 10 at least
X	Chem. Abs. 116:42907	1 & 10 at least
X	Chem. Abs. 115:115251	1 & 10 at least
X	Chem. Abs. 113:192043	1 & 10 at least
X	Chem. Abs. 113:79530	1 & 10 at least

X Document indicating lack of novelty or inventive step
Y Document indicating lack of inventive step if combined with one or more other documents of same category.

A Document indicating technological background and/or state of the art.
P Document published on or after the declared priority date but before the filing date of this invention.
E Patent document published on or after, but with priority date earlier

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